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(72) Inventors: CRITCHFIELD, James, E.; 5017 Placid Place, Austin, TX 78731 (US). MARQUIS, Edward, T.; 9004 Collinfield Drive, Austin, TX 78758 (US).

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(54) Title: LANDFILL GAS TREATMENT WITH PROPYLENE CARBONATE

(57) Abstract

This invention concerns a process useful for removing carbon dioxide from landfill gas, comprising: contacting the landfill gas which contains carbon dioxide, nitrogen, methane, and oxygen with alkylene carbonate under conditions effective to remove at least a portion of the carbon dioxide from the stream. The landfill gas may also contain chlorinated hydrocarbons, which may also have a detrimental effect on use of amines. Advantageously, use of alkylene carbonate may be used successfully despite the presence of both oxygen and chlorinated hydrocarbons.

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LANDFILL GAS TREATMENT WITH PROPYLENE CARBONATE

BACKGROUND OF INVENTION

This invention relates to a process for separation of carbon dioxide from a gaseous stream from a landfill which contains oxygen and/or chlorinated hydrocarbon, carbon dioxide and methane.

It is well known that landfills generate a substantial amount of methane. It would be highly desirable to separate the methane to thereby provide a source of this fuel. Amines are frequently used to sweeten sour gas withdrawn underground wells. However, a problem exists in that, unlike gas from underground wells, oxygen and other impurities such as chlorinated hydrocarbons are typically present in the gaseous stream which is collected from the landfill. Amines are prone to degradation in the presence of oxygen. Hence, amines do not provide a suitable absorbent composition for removing carbon dioxide from the landfill gas. A need thus exists for a workable carbon dioxide absorbent composition which is not prone to oxygen degradation for use in removing carbon dioxide from landfill gas. A solution to this need would be highly desirable.

SUMMARY OF INVENTION

In one broad respect, this invention is a process useful for removing carbon dioxide from landfill gas, comprising: contacting the landfill gas which contains carbon dioxide, methane, and oxygen with alkylene carbonate under conditions effective to remove at least a portion of the carbon dioxide from the stream. In another broad respect, this invention is a process useful for removing carbon dioxide from landfill gas, comprising: obtaining a gas stream from a landfill or dump which contains carbon dioxide, methane, and oxygen; contacting the gas stream alkylene carbonate under conditions effective to remove at least a portion of the carbon dioxide from the stream. The term landfill gas as used herein refers to gas obtained from landfills, dumps and the like. Thus, the term landfill gas refers to gas that

is being generated by the rotting, decay, biological break down of organic matter, degradation of dump materials and so forth. In addition to methane and or other hydrocarbons being generated by the landfill, the landfill gasses may contain chlorinated hydrocarbons such as methyl chloride, chloroform, chloroform and carbon tetrachloride, and higher carbon containing chlorinated hydrocarbons, which may also have a detrimental effect on use of amines. The amount of impurities such as chlorinated hydrocarbons and oxygen, individually or collectively, may be greater than about 1 percent, and in one embodiment may be greater than about 2 percent by volume of the landfill gas to be treated. Advantageously, use of alkylene carbonate may be used successfully despite the presence of both oxygen and chlorinated hydrocarbons. While it has been known to use propylene carbonate to remove carbon dioxide from natural gas from naturally occurring subterranean sources (see, for example, U.S. 2,926,751; U.S. 4,097,250; U.S. 4,449,994; and U.S. 4,749,555), these natural deposits did not contain oxygen and/or chlorinated hydrocarbons. For example, U.S. 4,097,250 describes a typical natural gas composition as containing methane, ethane, higher alkanes, nitrogen, carbon dioxide and optionally hydrogen sulfide in the case of an acidic gas source. These references are silent with respect to treatment of gas that also contains oxygen and/or chlorinated hydrocarbons. The present invention, by contrast, is directed specifically to washing of landfill gas that also contains oxygen and/or chlorinated hydrocarbons.

This invention has a number of advantages. Notably, alkylene carbonate such as propylene carbonate provides for excellent carbon dioxide removal from landfill gas. What is more, alkylene carbonate, propylene carbonate for instance, does not degrade to a significant degree in the presence of oxygen under absorbent conditions. Moreover, propylene carbonate serves to remove carbon dioxide without degrading or otherwise being rendered un-useful in the presence of chlorinated hydrocarbons. Thus, use of propylene carbonate as the absorbent solution provides a solution to the needs and disadvantages discussed above. Advantageously, the alkylene carbonate is useful for removal of carbon dioxide in order to improve the combustibility of landfill gas.

DETAILED DESCRIPTION OF THE INVENTION

The absorbent compositions of this invention include alkylene carbonate. The compositions may contain other components to enhance carbon dioxide removal, to provide corrosion resistance, and so forth.

The alkylene carbonate used in the present invention can contain from 2 to 10 carbon atoms. Representative examples of alkylene carbonates that may be employed in the practice of this invention include ethylene carbonate and propylene carbonate. In the practice of this invention, propylene carbonate is preferred.

The alkylene carbonate compositions may be used as the absorbent solution in conventional absorbent apparatus, such as stripping columns. For example, the alkylene carbonate may be used in place of the amine composition used in the stripping column and system as disclosed in U.S. 4,336,233, U.S. 4,449,994 and U.S. 4,749,555, incorporated herein by reference. In this regard, the alkylene carbonate solutions of this invention are used as absorbents to wash landfill gas.

The process may be run at conventional temperatures and pressures. Typically, the temperature may be as high as 100°C and is more typically less than about 65°C; may be as low as about 0 °C and more typically more than about 20 °C. Typically, pressure is atmospheric or superatmospheric. Contact times may vary depending on the temperature, initial carbon dioxide and/or chlorinated hydrocarbon concentration, desired carbon dioxide concentration in the exit stream, number of trays or packing in a column, and the like. This invention is preferably carried out in as a continuous process wherein landfill gas is continuously contacted with alkylene carbonate in a column. Conventional columns and related equipment may be used in the practice of this invention.

In one embodiment of this invention, the alkylene carbonate solution is contacted with the gas in a direction countercurrent to the flow of the landfill gas to be treated. For instance, the alkylene carbonate may be introduced into the top of the column as by spraying or misting

and allowed to descend as gas introduced at the bottom of the column flows upward. The alkylene carbonate may be sprayed in at the top of the column using conventional equipment with liquid alkylene carbonate saturated with carbon dioxide being collected at the base of the column. Alternatively, the landfill gas may be passed over a series of trays bearing the alkylene carbonate, using conventional methods. The contact of the alkylene carbonate and gas by such counterflow results in the uptake of the carbon dioxide into the liquid alkylene carbonate. Alternatively, cocurrent contacting may be used. As an alternative to use of a stripping column (or "tower"), the gas may be simply bubbled or sparged through a vessel containing the alkylene carbonate solution. If columns are employed in the process of this invention, the columns may be optionally packed or fitted with plates (trays).

The process of this invention may be carried out batchwise, intermittently or continuously. Likewise, the process can be repeated on effluent gas from a first process run to effect additional carbon dioxide removal. Similarly, the process can be run in stages with multiple columns or the like to achieve incremental removal of carbon dioxide as from the gas as it passes through multiple towers.

The alkylene carbonate composition laden with carbon dioxide may be readily regenerated as by heating with or without a separate or contemporaneous reduced pressure (e.g., flashing off of the carbon dioxide). Alternatively, the alkylene carbonate may be regenerated by use of a stripping gas which removes at least a portion of the carbon dioxide from the carbon dioxide laden alkylene carbonate composition. Such stripping gas can be any gas which has a partial pressure of carbon dioxide less than that of the carbon dioxide laden alkylene carbonate composition. For example, depending on the composition to be treated, the stripping gas may be air, nitrogen, argon, and so forth.

Some processes may be advantageously run whereby only a portion of the composition in a column is removed at any given time, regenerated, and recycled to the column. In this regard, the composition may be continuously regenerated by withdrawing a stream from the bottom of the column.

Further modifications and alternative embodiments of this invention will be apparent to those skilled in the art in view of this description. Accordingly, this description is to be construed as illustrative only and is for the purpose of teaching those skilled in the art the manner of carrying out the invention. It is to be understood that the forms of the invention herein shown and described are to be taken as illustrative embodiments. Equivalent elements or materials may be substituted for those illustrated and described herein, and certain features of the invention may be utilized independently of the use of other features, all as would be apparent to one skilled in the art after having the benefit of this description of the invention.

WHAT IS CLAIMED IS:

1. A process useful for removing carbon dioxide from landfill gas, comprising: contacting the landfill gas which contains carbon dioxide, methane, and oxygen with alkylene carbonate under conditions effective to absorb at least a portion of the carbon dioxide from the stream into the alkylene carbonate.

2. The process of claim 1 including the step of obtaining the landfill gas from a landfill or dump.

3. The process of any one of claims 1-2 wherein the alkylene carbonate is propylene carbonate.

4. The process of any of claims 1-3 wherein the contacting occurs in a stripping column.

5. The process of any of claims 1-4 wherein the contacting occurs in a stripping column containing packing or trays.

6. The process of any of claims 1-5 wherein the contacting occurs in countercurrent fashion in a stripping column.

7. The process of any of claims 1-6 further comprising regenerating alkylene carbonate laden with carbon dioxide and recycling the regenerated alkylene carbonate to the contacting step.

8. The process of any of claims 1-7 wherein the oxygen, chlorinated hydrocarbon or combination thereof is present in an amount greater than 1 percent by volume of the landfill gas.

9. The process of any of claims 1-8 wherein the landfill gas further comprises chlorinated hydrocarbons.

10. The process of any of claims 1-9 wherein the landfill gas contains at least 1 percent of oxygen, chlorinated hydrocarbon or combination thereof.

11. The process of any of claims 1-10 wherein regeneration of the alkylene carbonate is conducted by heating under reduced pressure.

12. The process of any of claims 1-11 wherein the contacting occurs in a column containing packing or trays.

13. The process of any of claims 1-12 wherein the contacting occurs at a temperature less than about 65 degrees Centigrade.

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(72) Inventors: CRITCHFIELD, James, E.; 5017 Placid Place, Austin, TX 78731 (US). MARQUIS, Edward, T.; 9004 Collinfield Drive, Austin, TX 78758 (US).

(74) Agent: O'KEEFE, Robert, M.; Jones, O'Keefe, Egan & Peterman, Building C, Suite 200, 1101 Capital of Texas Highway South, Austin, TX 78746 (US).

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(57) Abstract

This invention concerns a process useful for removing carbon dioxide from landfill gas, comprising: contacting the landfill gas which contains carbon dioxide, nitrogen, methane, and oxygen with alkylene carbonate under conditions effective to remove at least a portion of the carbon dioxide from the stream. The landfill gas may also contain chlorinated hydrocarbons, which may also have a detrimental effect on use of amines. Advantageously, use of alkylene carbonate may be used successfully despite the presence of both oxygen and chlorinated hydrocarbons.

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INTERNATIONAL SEARCH REPORT

Inter nal Application No
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A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 B01D53/14 C10L3/10 C07C7/11

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 6 B01D C10L C07C B09B

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 2 926 751 A (KOHL ET AL.) 1 March 1960 (1960-03-01) cited in the application	1-8, 10-13
Y	column 1, line 14 - line 18; claim 1	9
Y	EP 0 180 670 A (CRYOTEC ENERGY SYSTEMS CO LTD) 14 May 1986 (1986-05-14) page 1, line 3 - line 16 page 2, line 16 - line 21 page 4, line 21 - line 29 page 5, line 28 -page 6, line 10	9

☐ Further documents are listed in the continuation of box C.

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Date of the actual completion of the international search	Date of mailing of the international search report
23 March 2000	31/03/2000

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INTERNATIONAL SEARCH REPORT

information on patent family members

Inter nal Application No
PCT/US 99/02567

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
US 2926751 A	01-03-1960	GB 893423 A	
EP 0180670 A	14-05-1986	NONE	

LANDFILL GAS TREATMENT WITH PROPYLENE CARBONATE

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Inventor(s): CRITCHFIELD JAMES E; MARQUIS EDWARD T
Applicant(s):: HUNTSMAN SPEC CHEM CORP (US)

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Priority Number(s): US19980073919P 19980206
IPC Classification: B01J
EC Classification: B01D53/62
Equivalents: AU2589199

Abstract

This invention concerns a process useful for removing carbon dioxide from landfill gas, comprising: contacting the landfill gas which contains carbon dioxide, nitrogen, methane, and oxygen with alkylene carbonate under conditions effective to remove at least a portion of the carbon dioxide from the stream. The landfill gas may also contain chlorinated hydrocarbons, which may also have a detrimental effect on use of amines. Advantageously, use of alkylene carbonate may be used successfully despite the presence of both oxygen and chlorinated hydrocarbons.

The alkylene carbonate used in the present invention can contain from 2 to 10 carbon atoms. Representative examples of alkylene carbonates that may be employed in the practice of this invention include ethylene carbonate and propylene carbonate. In the practice of this invention, propylene carbonate is preferred.

The alkylene carbonate compositions may be used as the absorbent solution in conventional absorbent apparatus, such as stripping columns. For example, the alkylene carbonate may be used in place of the amine composition used in the stripping column and system as disclosed in U.S. 4,336,233, U.S. 4,449,994 and U.S. 4,749,555, incorporated herein by reference. In this regard, the alkylene carbonate solutions of this invention are used as absorbents to wash landfill gas.

The process may be run at conventional temperatures and pressures. Typically, the temperature may be as high as 100"C and is more typically less than about 65"C; may be as low as about 0 "C and more typically more than about 20 C. Typically, pressure is atmospheric or superatmospheric. Contact times may vary depending on the temperature, initial carbon dioxide and/or chlorinated hydrocarbon concentration, desired carbon dioxide concentration in the exit stream, number of trays or packing in a column, and the like. This invention is preferably carried out in as a continuous process wherein landfill gas is continuously contacted with alkylene carbonate in a column. Conventional columns and related equipment may be used in the practice of this invention.

In one embodiment of this invention, the alkylene carbonate solution is contacted with the gas in a direction countercurrent to the flow of the landfill gas to be treated. For instance, the alkylene carbonate may be introduced into the top of the column as by spraying or misting and allowed to descend as gas introduced at the bottom of the column flows upward. The alkylene carbonate may be sprayed in at the top of the column using conventional equipment with liquid alkylene carbonate saturated with carbon dioxide being collected at the base of the column. Alternatively, the landfill gas may be passed over a series of trays bearing the alkylene carbonate, using conventional methods. The contact of the alkylene carbonate and gas by such counterflow results in the uptake of the carbon dioxide into the liquid alkylene carbonate. Alternatively, cocurrent contacting may be used. As an alternative to use of a stripping column (or "tower"), the gas may be simply bubbled or sparged through a vessel containing the alkylene carbonate solution. If columns are employed in the process of this invention, the columns may be optionally packed or fitted with plates (trays).

The process of this invention may be carried out batchwise, intermittently or continuously. Likewise, the process can be repeated on effluent gas from a first process run to effect additional carbon dioxide removal. Similarly, the process can be run in stages with multiple columns or the like to achieve incremental removal of carbon dioxide as from the gas as it passes through multiple towers.

The alkylene carbonate composition laden with carbon dioxide may be readily regenerated as by heating with or without a separate or contemporaneous reduced pressure (e.g., flashing off of the carbon dioxide). Alternatively, the alkylene carbonate may be regenerated by use of a stripping gas which removes at least a portion of the carbon dioxide from the carbon dioxide laden alkylene carbonate composition. Such stripping gas can be any gas which has a partial pressure of carbon dioxide less than that of the carbon dioxide laden alkylene carbonate composition. For example, depending on the composition to be treated, the stripping gas may be air, nitrogen, argon, and so forth.

Some processes may be advantageously run whereby only a portion of the composition in a column is removed at any given time, regenerated, and recycled to the column. In this regard, the composition may be continuously regenerated by withdrawing a stream from the bottom of the column.

Further modifications and alternative embodiments of this invention will be apparent to those skilled in the art in view of this description. Accordingly, this description is to be construed as illustrative only and is for the purpose of teaching those skilled in the art the manner of carrying out the invention. It is to be understood that the forms of the invention herein shown and described are to be taken as illustrative embodiments. Equivalent elements or materials may be substituted for those illustrated and described herein, and certain features of the invention may be utilized independently of the use of other features, all as would be apparent to one skilled in the art after having the benefit of this description of the invention.

Claims

WHAT IS CLAIMED IS: 1. A process useful for removing carbon dioxide from landfill gas, comprising: contacting the landfill gas which contains carbon dioxide, methane, and oxygen with alkylene carbonate under conditions effective to absorb at least a portion of the carbon dioxide from the stream into the alkylene carbonate.

2. The process of claim 1 including the step of obtaining the landfill gas from a landfill or dump.

3. The process of any one of claims 1-2 wherein the alkylene carbonate is propylene carbonate.

4. The process of any of claims 1-3 wherein the contacting occurs in a stripping column.

5. The process of any of claims 1-4 wherein the contacting occurs in a stripping column containing packing or trays.

6. The process of any of claims 1-5 wherein the contacting occurs in countercurrent fashion in a stripping column.

7. The process of any of claims 1-6 further comprising regenerating alkylene carbonate laden with carbon dioxide and recycling the regenerated alkylene carbonate to the contacting step.

8. The process of any of claims 1-7 wherein the oxygen, chlorinated hydrocarbon or combination thereof is present in an amount greater than 1 percent by volume of the landfill gas.

9. The process of any of claims 1-8 wherein the landfill gas further comprises chlorinated hydrocarbons.

10. The process of any of claims 1-9 wherein the landfill gas contains at least 1 percent of oxygen, chlorinated hydrocarbon or combination thereof.

11. The process of any of claims 1 - 10 wherein regeneration of the alkylene carbonate is conducted by heating under reduced pressure.

12. The process of any of claims 1-11 wherein the contacting occurs in a column containing packing or trays.

13. The process of any of claims 1-12 wherein the contacting occurs at a temperature less than about 65 degrees Centigrade.

Description

LANDFILL GAS TREATMENT WITH PROPYLENE CARBONATE
BACKGROUND OF INVENTION
This invention relates to a process for separation of carbon dioxide from a gaseous stream from a landfill which contains oxygen and/or chlorinated hydrocarbon, carbon dioxide and methane.

It is well known that landfills generate a substantial amount of methane. It would be highly desirable to separate the methane to thereby provide a source of this fuel. Amines are frequently used to sweeten sour gas withdrawn underground wells. However, a problem exists in that, unlike gas from underground wells, oxygen and other impurities such as chlorinated hydrocarbons are typically present in the gaseous stream which is collected from the landfill. Amines are prone to degradation in the presence of oxygen. Hence, amines do not provide a suitable absorbent composition for removing carbon dioxide from the landfill gas. A need thus exists for a workable carbon dioxide absorbent composition which is not prone to oxygen degradation for use in removing carbon dioxide from landfill gas. A solution to this need would be highly desirable.

SUMMARY OF INVENTION
In one broad respect, this invention is a process useful for removing carbon dioxide from landfill gas, comprising: contacting the landfill gas which contains carbon dioxide, methane, and oxygen with alkylene carbonate under conditions effective to remove at least a portion of the carbon dioxide from the stream. In another broad respect, this invention is a process useful for removing carbon dioxide from landfill gas, comprising: obtaining a gas stream from a landfill or dump which contains carbon dioxide, methane, and oxygen; contacting the gas stream alkylene carbonate under conditions effective to remove at least a portion of the carbon dioxide from the stream. The term landfill gas as used herein refers to gas obtained from landfills, dumps and the like. Thus, the term landfill gas refers to gas that is being generated by the rotting, decay, biological break down of organic matter, degradation of dump materials and so forth. In addition to methane and or other hydrocarbons being generated by the landfill, the landfill gasses may contain chlorinated hydrocarbons such as methyl chloride, chloroform, chloroform and carbon tetrachloride, and higher carbon containing chlorinated hydrocarbons, which may also have a detrimental effect on use of amines. The amount of impurities such as chlorinated hydrocarbons and oxygen, individually or collectively, may be greater than about 1 percent, and in one embodiment may be greater than about 2 percent by volume of the landfill gas to be treated. Advantageously, use of alkylene carbonate may be used successfully despite the presence of both oxygen and chlorinated hydrocarbons. While it has been known to use propylene carbonate to remove carbon dioxide from natural gas from naturally occurring subterranean sources (see, for example, U.S. 2,926,751; U.S. 4,097,250; U.S. 4,449,994; and U.S. 4,749,555), these natural deposits did not contain oxygen and/or chlorinated hydrocarbons. For example, U.S.

4,097,250 describes a typical natural gas composition as containing methane, ethane, higher alkanes, nitrogen, carbon dioxide and optionally hydrogen sulfide in the case of an acidic gas source. These references are silent with respect to treatment of gas that also contains oxygen and/or chlorinated hydrocarbons. The present invention, by contrast, is directed specifically to washing of landfill gas that also contains oxygen and/or chlorinated hydrocarbons.

This invention has a number of advantages. Notably, alkylene carbonate such as propylene carbonate provides for excellent carbon dioxide removal from landfill gas. What is more, alkylene carbonate, propylene carbonate for instance, does not degrade to a significant degree in the presence of oxygen under absorbent conditions. Moreover, propylene carbonate serves to remove carbon dioxide without degrading or otherwise being rendered un-useful in the presence of chlorinated hydrocarbons. Thus, use of propylene carbonate as the absorbent solution provides a solution to the needs and disadvantages discussed above.

Advantageously, the alkylene carbonate is useful for removal of carbon dioxide in order to improve the combustibility of landfill gas.

DETAILED DESCRIPTION OF THE INVENTION
The absorbent compositions of this invention include alkylene carbonate. The compositions may contain other components to enhance carbon dioxide removal, to provide corrosion resistance, and so forth.